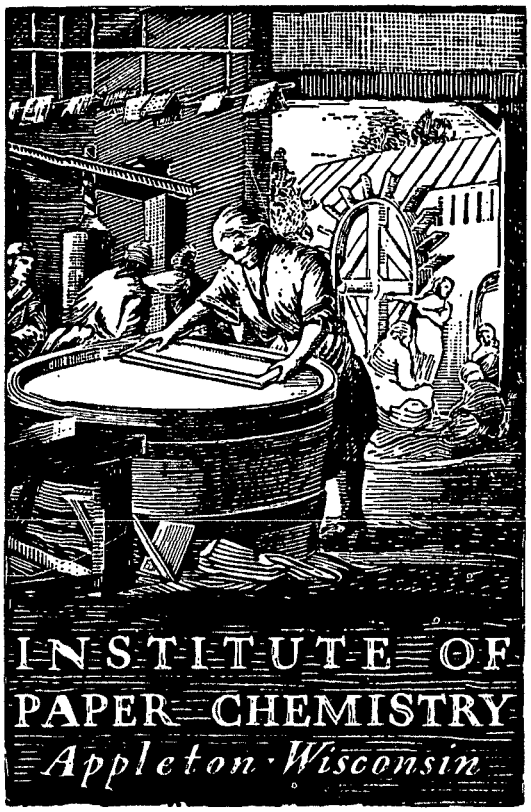

SUSPENDED SOLIDS REMOVAL

Project 3352

Report One

A Progress Report

to

MEMBERS OF THE INSTITUTE OF PAPER CHEMISTRY

January 30, 1979

THE INSTITUTE OF PAPER CHEMISTRY

Appleton, Wisconsin

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SUSPENDED SOLIDS REMOVAL

SUMMARY

The removal of suspended solids from in-plant streams or secondary effluents is important in view of the proposed 1983 EPA guidelines. This project was, therefore, undertaken to study four commonly used solid-liquid separation processes: granular media filtration, dissolved air flotation, microscreening and sedimentation. In the past century, considerable improvement has been made to increase the efficiency of these clarification processes. We believe that any further improvement will come primarily from better understanding of the separation mechanisms. This progress report covers work done on granular media (sand) filtration.

Basic separation mechanisms affecting the removal of suspended solids in media filtration are investigated by separating the effects of hydrodynamic and electrokinetic forces. To date, our results show that particles in the size range of 0.1 to 5 μm are difficult to remove. The separation improves considerably when one adjusts pH or increases ionic strength or adds a coagulation aid to destabilize colloidal particles. The presence of starch reduces filtration velocity significantly.

The effectiveness of granular media filtration in treating clarified water from saveall units and from biological treatment systems (secondary effluent) is being investigated in light of the separation mechanisms proposed here. This, together with our work on dissolved air flotation, will be discussed in the next report.

INTRODUCTION

Stringent effluent regulations followed by strict enforcement have encouraged many industries to modify their processes and/or improve their treatment facilities. The pulp and paper industry has made significant progress to reduce the consumption of freshwater and improve the quality of discharged effluent. One of the problems in closing the mill water cycle is the control of suspended solids, especially in the size range of 0.1 to 5 μm. For example, when the white water cycle is closed in a paper mill, suspended solids clog wires, felts and shower nozzles (1). Furthermore, buildup of suspended solids and fiber fines can affect the drainage rate and the quality of the finished product. The removal and control of suspended solids is thus quite important to the pulp and paper industry.

Solid-liquid separation processes, like sedimentation, filtration, and flotation, have been in use in the pulp and paper industry, as well as elsewhere, to remove suspended solids. When these processes are used for white water treatment, they are collectively known as saveall processes in the paper industry. In the past century, considerable improvement has been made to increase the efficiency of these clarification processes. We believe any further improvement will come primarily from better understanding of the separation mechanisms.

OBJECTIVE

The overall objective of this project is to reduce the discharge of suspended solids from pulp and paper mill effluents by advanced treatment and/or process control. This broad objective can be achieved through better understanding of commonly used solid-liquid separation processes, namely,

1. granular media filtration
2. dissolved air flotation
3. microscreening
4. sedimentation

Various separation mechanisms which affect the efficiency of these processes will be studied. This will help us to select a separation process or modify the existing one in a systematic way. Our work on granular media (sand) filtration is presented in this report.

Some potentially useful processes like centrifugation, reverse osmosis, ultrafiltration, and freezing are not considered at this time since they are not economical compared to the above four methods. If the economic picture improves or needs change, we will investigate such processes.

APPROACH

Suspended solids usually encountered in papermaking operations (e.g., titanium dioxide, latex, and clay) were used for the initial phase of this program. These particles range from 0.1 to 5 μm. Particles of this size are, in general, difficult to remove. Particles much smaller than 0.1 μm, due to their intense Brownian motion, and particles much larger than 5 μm, due to their greater settling velocity, are relatively easier to remove.

There are several exceptions to this general rule simply because separation of colloidal particles is quite complicated and depends on particle charge, density, etc., in addition to particle size. Nevertheless, the removal of particles in this difficult-to-remove size range of 0.1 to 5 μm should provide a stringent test for the selection of a separation method.

Even though granular media filtration has been used in water treatment for over a century, considerable progress has been made only recently in understanding

various separation mechanisms. A brief review of recent developments is presented in the next section. In the subsequent sections our results are presented and examined in light of the current developments.

GRANULAR MEDIA FILTRATION

Granular media filtration is widely used for separation of suspended solids. It is generally operated on a batch or semicontinuous mode by using a fixed bed of sand grains or similar material. Although some continuous sand filtration units are in operation, they are not yet popular.

Granular media filtration can be divided into two categories: cake filtration and depth filtration (2-5). Pore openings in granular beds are such that particles greater than 30 μm will generally not pass through. These particles will, therefore, accumulate at the bed surface, forming a cake. Further separation is effected by this cake, and consequently it is called cake filtration. Particles smaller than 3 μm, on the other hand, are separated by deposition on the grains throughout the bed depth. Thus, separation of smaller particles is termed depth filtration. Both pore blockage and surface deposition are operative for particles from 3 to 30 μm.

In this project we are primarily concerned with depth filtration. Since cake filtration is somewhat similar to microscreening, we will consider these two together in future work.

RECENT DEVELOPMENTS IN DEPTH FILTRATION

Separation of colloidal particles in granular media filtration is believed to occur in two steps (5-8):

1. transport of particles from the bulk fluid phase to the vicinity of grains and,
2. attachment of the particles to the grain surface.

The transport step is primarily influenced by the prevailing hydrodynamic conditions and depends on particle size, grain size, filtration velocity, particle density, and temperature (as it affects fluid viscosity and Brownian diffusion).

The attachment step is electrochemical. The adhesion of particle to grain will depend on their electric charges. If particles and grains have opposite charges, this may promote adhesion. When both particles and grains have a charge of the same sign, the degree of adhesion will be determined by a net balance between Van der Waals attractive forces and double layer repulsive forces. [Note: Van der Waals attractive forces are always present, and they are independent of the charge on the particles. Increasing the number of atoms per unit volume in each particle, i.e., increasing particle density, increases the magnitude of these attractive forces (9).] The charge on colloidal particles can be varied by changing pH and/or ionic strength.

Separation mechanisms encountered in depth filtration are somewhat similar to those found in the papermaking operation. Dyer (10) used this idea to study retention mechanisms in the deposition of colloidal particles on cylindrical fibers. More work in this direction is needed to improve our understanding of retention mechanisms.

Before proceeding further with the review of recent developments, let us define some of the common terms used in the filtration literature. The filtration coefficient, λ, introduced by Iwasaki, in 1937 (11), is still used today.

Iwasaki proposed that the rate of removal of colloidal particles per unit depth of filter is proportional to the local concentration:

$$\frac{dC}{dZ} = - \lambda C$$

where C is the concentration of suspended solids at any time and depth, and Z is the length of the bed measured from the top. The filtration coefficient, λ, is assumed to be independent of Z. The above equation can be integrated to give

$$\ln \frac{C_o}{C_L} = \lambda L$$

where C_o is the feed concentration, C_L is the concentration of suspended solids at the end of the bed, and L is the height of the bed (see Fig. 1).

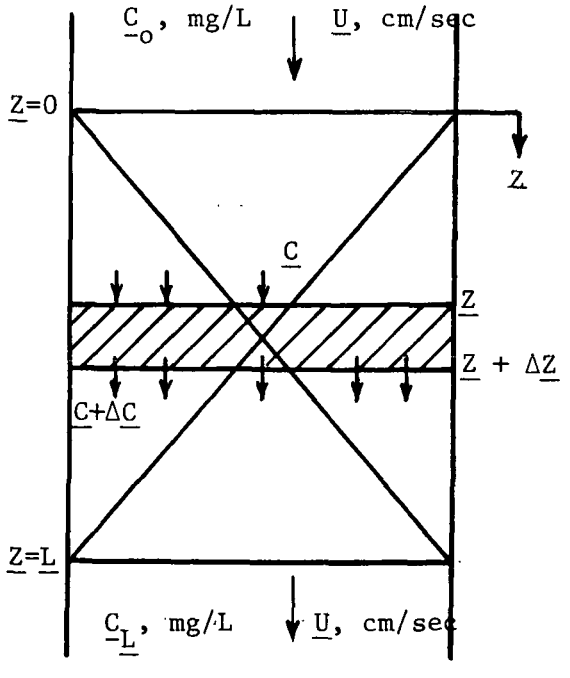

Figure 1. Schematic Diagram of Granular Media Filtration Bed

Let us now consider the mass balance over an extremely thin layer, thickness ΔZ, of a bed (7):

No. of particles entering the slice in unit time = $U \cdot A \cdot C$

No. of particles leaving the slice in unit time = $U \cdot A \cdot (C + \Delta C)$

No. of particles removed in unit time = $-U \cdot A \cdot \Delta C$

In the above expressions, U is the superficial velocity and A is the cross-sectional area of the bed, and ΔC represents change in particle concentration of fluid due to passage through the infinitesimal bed element (see Fig. 1). Note that ΔC is negative since particles are removed from the fluid.

$$\text{No. of grains in the slice} = \frac{A \cdot \Delta Z(1-\varepsilon)}{\left(\frac{\pi}{6} d_g^3\right)}$$

where ε is the bed porosity and d_g is the average diameter of grains.

$$\text{No. of particles encountered by each grain in unit time} = U \cdot C \cdot \left(\frac{\pi}{4} d_g^2\right)$$

Let us now define collision efficiency and collection efficiency:

$$\text{Collision Efficiency} = \eta = \frac{\text{No. of particles colliding with a grain}}{\text{No. of particles encountered by a grain}}$$

$$\text{Collection Efficiency} = \alpha = \frac{\text{No. of particles adhering to a grain}}{\text{No. of particles colliding with a grain}}$$

As explained earlier, collision efficiency will be influenced by the hydrodynamic conditions, whereas collection efficiency will be determined by the charge on colloid particles and grains and by the magnitude of the attractive forces. From these definitions we have:

$$\text{No. of particles collected by each grain in unit time} = U \cdot C \cdot \frac{\pi}{6} d_g^2 \cdot \eta \cdot \alpha$$

Since we know the number of grains in the slice, we can write

No. of particles removed in the slice in unit time

$$= \frac{A \cdot \Delta Z(1-\varepsilon)U \cdot C \cdot}{\frac{\pi}{6} d_g^3} \left(\frac{\pi}{4} d_g^2\right) \eta \cdot \alpha$$

$$= \left(\frac{3}{2d_g}\right) A \cdot \Delta Z(1-\varepsilon)U \cdot C \cdot \eta \cdot \alpha$$

Equating the two expressions for the number of particles removed in the slice in unit time we get:

$$-U \cdot A \cdot \Delta C \cdot = \left(\frac{3}{2d_g}\right) A \cdot \Delta Z(1-\varepsilon)U \cdot C \cdot \eta \cdot \alpha$$

Rearranging this equation and taking limits as ΔC and ΔZ approach zero one gets:

$$-\frac{dC}{dZ} = \left(\frac{3}{2d_g}\right) \eta \cdot \alpha(1-\varepsilon)C$$

Integration yields

$$\ln\left(\frac{C_o}{C_L}\right) = \left(\frac{3}{2d_g}\right) \eta \cdot \alpha \ (1-\varepsilon)L$$

Clearly, λ, η, and α are related:

$$\lambda = \frac{3}{2d_g} \eta \cdot \alpha(1-\varepsilon)$$

In order to predict filtration performance, it is necessary to relate λ or η and α in terms of the properties of the suspension, and grains and operating conditions (velocity and temperature). Many elaborate mathematical models (e.g., 12-15) have been developed to obtain such relations. However, agreement with experimental data is not satisfactory, and therefore only some of the important models will be discussed here.

It appears from the literature that there are basically four ways to transport suspended particles to a grain (see Fig. 2):

(1) Brownian diffusion: As a colloidal particle undergoes random Brownian motion while following a streamline, it may collide with a grain. The random motion can be described by a diffusion equation, with diffusion coefficient approximated by the Stokes-Einstein relation:

$$D = \frac{kT}{3\pi\mu d}$$

where

$\underline{D}$ = diffusion coefficient, cm^2/sec

$\underline{k}$ = Boltzmann constant = 1.38×10^{-6} $\frac{erg}{molecule\ °K}$ $\left(= \frac{g\ cm^2}{sec^2\ °K}\right)$

$\underline{T}$ = absolute temperature, °K

μ = viscosity of the fluid, poise

$\underline{d}$ = diameter of the particle, cm

(2) Sedimentation: Due to density differences between a particle and a fluid, the particle may deviate from a streamline under the action of gravity. Thus, in the case of downward flow, collision efficiency of particles heavier than water may increase (see Fig. 2).

(3) Interception: As a particle follows a streamline, it may come in contact with a grain by virtue of its size.

(4) Straining: Colloidal particles will contact grains when they encounter constriction or dead space in a packed bed.

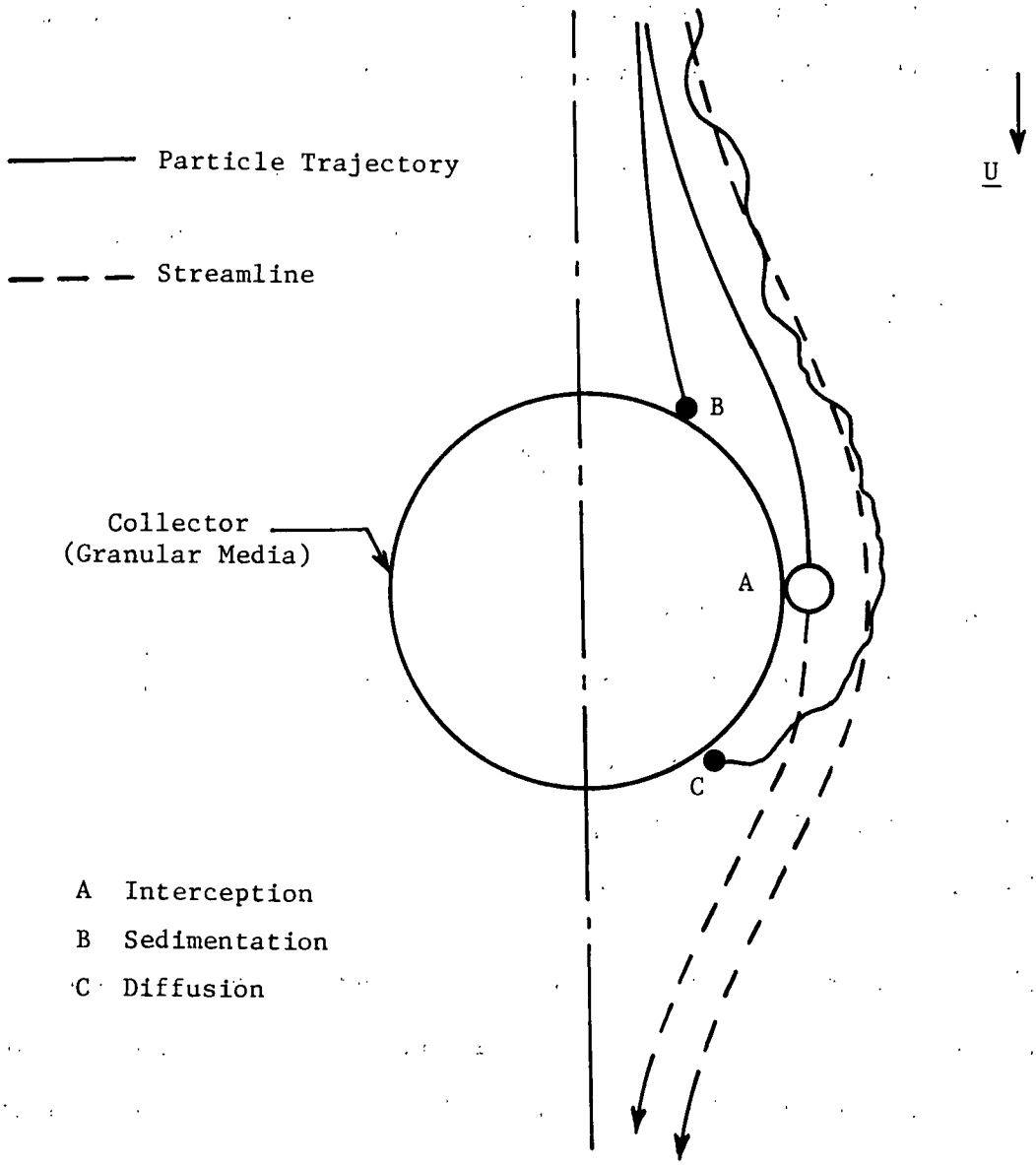

Figure 2. Modes of Action of the Basic Transport Mechanisms (7)

From the information available in the literature, the following conclusions can be drawn about the magnitude and relative importance of these transport modes in a given situation (4,6,12-15):

- Collision efficiency due to Brownian diffusion alone can be given by:

$$\eta_D = A_D(Pe)^{-2/3} = \frac{A_D}{(3\pi)^{2/3}}\left(\frac{kT}{Ud_g\, d}\right)^{2/3}$$

where

A_D = a constant

Pe = Peclet number = Ud_g/D

The numerical constant A_D is about 4 if a Stokes velocity profile is assumed, whereas it depends on porosity (for $\varepsilon = 0.4$, $A_D \simeq 16$) when the equivalent cell model is used (16). Yao, et al. (6) pointed out that actual value may be somewhere between these two extremes.

- Contribution of sedimentation to the collision efficiency can be expressed as:

$$\eta_G = \frac{(\rho_p - \rho)g\, d^2}{18\, \mu U}$$

where

ρ_p = density of particle

ρ = fluid density

g = acceleration due to gravity

- The importance of collision by interception is not clear from the literature. If parabolic velocity profile is assumed, one gets

$$\eta_I = A_I\left(\frac{d}{d_g}\right)^2$$

On the other hand, assuming plug flow yields:

$$\eta_I = A_I\left(\frac{d}{d_g}\right)$$

• Straining will be important for particles larger than about 10 μm, and therefore it has not been considered in the analysis of depth filtration.

• Yao, et al. (6) solved the convective diffusion equation numerically to calculate the collision efficiency due to diffusion, sedimentation, and interception. Their results, presented in Fig. 3, show that:

(1) Brownian diffusion is important for small particles, d <1 μm, whereas sedimentation becomes important as particle diameter increases above 1 μm.

(2) Interception, as expressed by the square of the diameter ratio, is negligible compared to sedimentation.

(3) Contributions from diffusion, sedimentation, and interception are additive,

$$\eta = \eta_D + \eta_G + \eta_I$$

(4) Particles in the 1 μm range have the lowest collision efficiency for the parameters listed in Fig. 2.

Information on calculating collection efficiency or attachment efficiency, α, is not available in the literature. However, the above approach of separating the hydrodynamic and electrochemical effects by introducing η and α have been criticized by many investigators (12-15). It is argued that, as a colloid particle approaches a grain, the particle trajectory will be influenced by electrochemical forces, and consequently interaction between hydrodynamic and electrokinetic forces

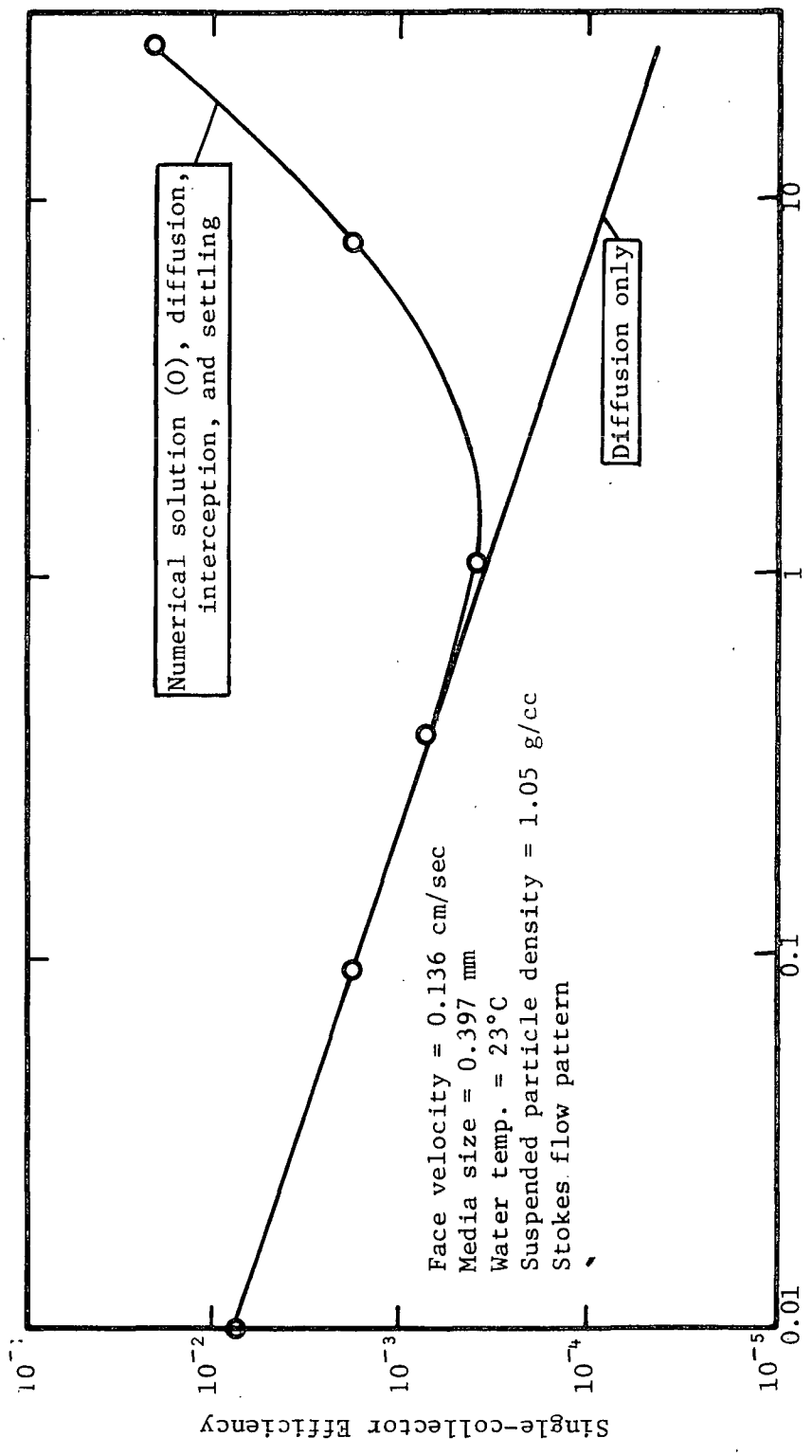

Figure 3. Theoretical Particle Transport Efficiency in a Porous Medium

should be taken into account. This complicates the analysis significantly, without noticeably improving the prediction. A review of these analyses is therefore not presented here, and the reader can refer to the original articles (12-15), review articles (17,18) and books (2-4).

Some researchers have followed a more or less empirical approach (19,20) to develop relationships between the filter coefficient λ and system parameters, ε, d, d_g U, μ, T or D, ρ and ρ_p. In view of the complicated nature of the particle-grain interaction and the large number of parameters, it is not surprising to note that such an approach has not been successful.

In this project, we plan to compromise between the theoretical and empirical approaches by developing a semitheoretical model. Results from our experiments, presented in the next section, will be analyzed in light of the existing and proposed models.

APPLICATIONS IN PULP AND PAPER INDUSTRY

If the proposed 1983 guideline for the amount of suspended solids discharged from mill effluents is enforced, the pulp and paper industry (like most other industries and municipal plants) will have to look for ways to further upgrade their effluents. Granular media filtration is ideal for such an advanced treatment (tertiary treatment).

EPA recommends media filtration to reduce suspended solids from 30 to 100 mg/L (normal in secondary effluent) to less than 10 mg/L. However, recently, Biskner, et al. (21) found that media filtration is not as effective in treating pulp and paper mill secondary effluent as it is in treating municipal secondary effluents because of the considerable difference in the particle size distribution.

Biskner, et al. found that particles in the mill secondary effluent are less than 10 μm, averaging around 2 μm. (As explained earlier, particles in the 0.1 to 5 μm range are generally difficult to remove and are separated by depth filtration.) On the other hand, particles in the municipal effluent have two size ranges, one similar to that in mill effluent and the other in the 20 to 200 μm range. The larger particles are removed by cake filtration. In view of the different separation mechanisms, it is not possible to extrapolate results from municipal treatment to pulp and paper mill waste water treatment.

Media filtration can be used not only for advanced treatment but it can also be used in the treatment of raw water and for in-plant control. The Kingsport mill of Mead Papers has been using media filtration to treat raw water since 1918 (22). Davis, et al. (23) report that in the Miquon mill of Weyerhaeuser Company, they were able to reduce the freshwater requirement by using media filtration and recycling the filtrate.

EXPERIMENTAL

PLAN

White water treatment was emphasized in this phase of the project. Therefore, separation of colloidal particles encountered in white water, namely, titanium dioxide, clay, and latex were considered first. These particles are in the difficult-to-remove size range of about 0.1 to 5 μm. Separation of these particles from single component suspensions and mixtures were considered at different ionic strengths and pH's. Also the separation of alum coagulated suspensions was studied. The filtration of fiber fines will be discussed in the next report.

It has been found that when "broke" containing starch is added to the stock, it affects the retention properties. In order to get further insight into this, we studied separation of the aforementioned particles in the presence of starch at various ionic strengths and pH and in the presence of alum.

EXPERIMENTAL SET-UP

The purpose of the sand filtration experiment was to measure filtration performance under various operating conditions.

A sand bed of 7.62 cm (3 inches) height and 2.54 cm (1 inch) was formed in a tube. A 200-mesh screen was placed at the bottom of the tube to hold the sand in place. Sand which passed through the 60-mesh screen (0.250 mm opening) and was retained by the 100-mesh screen (0.149 mm opening) was used. The average sand grain diameter, measured microscopically, was about 0.2 mm.

The flow diagram is shown in Fig. 4. Feed entered the sand bed through a constant head tank. A pool of liquid was maintained at the top of the bed to damp the impact of the incoming feed which otherwise will disrupt the bed. Flow rate through the bed was measured by a stopwatch and a graduated cylinder. Pressure drop across the bed was measured by a U-tube manometer containing tetrabromoethane (sp. gr. = 2.96).

Concentration of suspended solids in feed and filtrate was determined by filtration of a known volume of sample through a 0.45 μm filter. The following properties of suspensions were also measured:

1. particle size using a Coulter counter
2. zeta potential by electrophoresis using a Zeta meter
3. conductivity
4. pH

In addition, some of the samples were used to measure particle density using a pycnometer.

The porosity of the bed was found to be about 0.4. Pressure drop across the bed, for example, was 26.67 cm (10.5 inches) of tetrabromoethane when the flow rate of water at 20°C was 89.0 mL/min. Pressure drop calculated from the Kozeny-Carman equation (24,25)

$$\Delta P = \frac{U \cdot \mu L \cdot}{K \cdot}$$

was found to be about 11.6 inches, within 10% of the measured value. In the above equation ΔP represents the pressure drop across the bed of length L when a fluid with viscosity μ flows with a velocity U. The Kozeny constant K is related to sand grain diameter d and porosity ε:

$$K = \frac{d^2 \varepsilon^3}{180(1-\varepsilon)^2}$$

An error of 10% in the pressure drop measurement is considered quite good, since a number of approximations have been made in deriving the Kozeny-Carman equation.

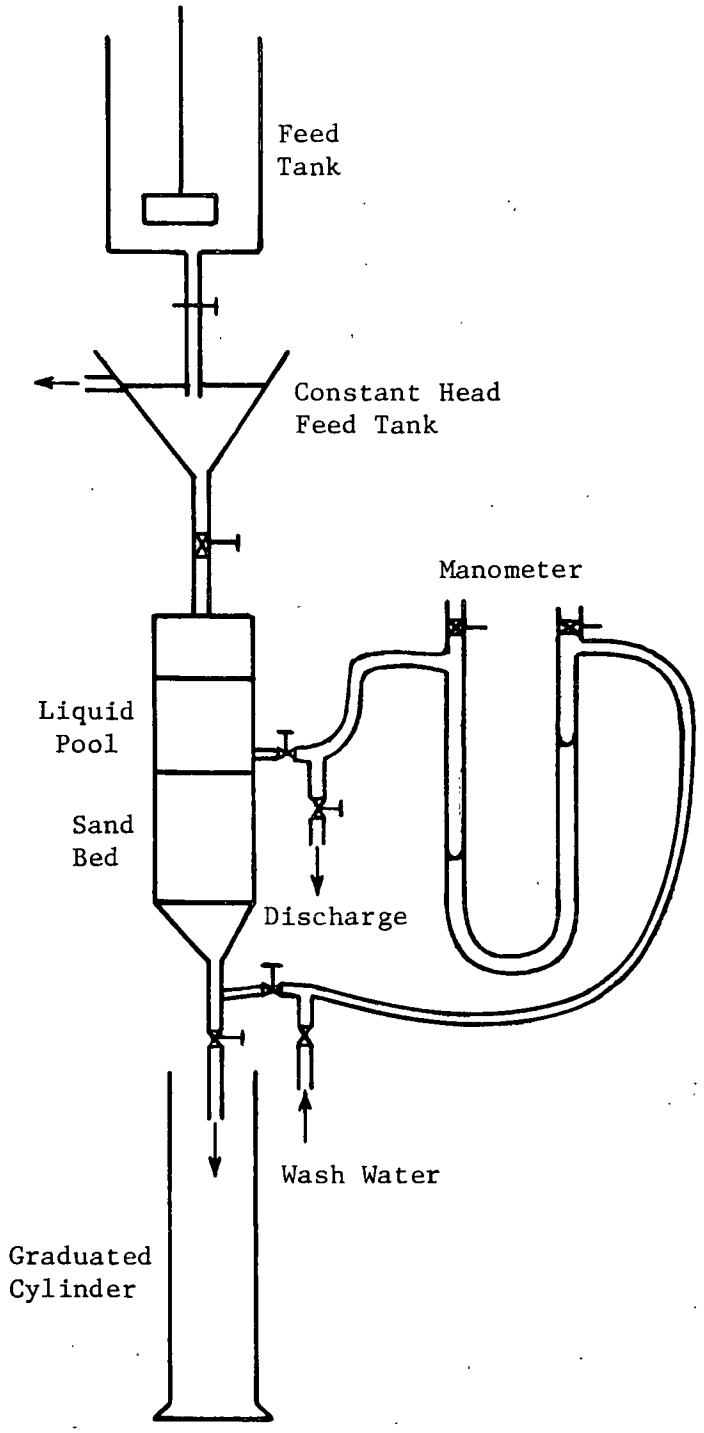

Figure 4. Experimental Set-up for Sand Filtration

RESULTS AND DISCUSSION

TiO_2 AND CLAY — DIFFERENT pH's AND IONIC STRENGTHS

Separation of titanium dioxide and clay particles by sand filtration was studied. Particle density and average particle diameter are presented in Table I. These particles are relatively heavy and are in the difficult-to-remove size range. Some experiments were carried out with latex particles which are considerably lighter than clay or titanium dioxide.

TABLE I

PHYSICAL PROPERTIES OF PARTICLES

	Density, g/cm^3	Diameter, μm
Titanium dioxide, (Anatase Zopaque RG)	4.0	0.4
No. 2 Grade, KCS (kaolin coating clay)	2.5	1.4
Latex	1.08	0.5

The effects of pH and ionic strength on separation of titanium dioxide and clay are shown in Table II. In these experiments, the flow was from a constant head tank and no attempt was made to control the flow rate. Filtrate velocity varied between 80 and 105 mL/min, except in one case it was quite low, 52.6 mL/min. Feed solids were generally kept at around 100 mg/L.

In three cases, filtrate concentration was not measurable. Since accuracy in weighing is about 0.2 mg and a 250 mL sample was taken, the error in measurement could be 0.8 mg/L. Therefore, the minimum concentration in the filtrate is assumed to be 0.8 mg/L.

TABLE II

SEPARATION OF TiO_2 AND CLAY AT DIFFERENT pH's AND IONIC STRENGTHS

pH	NaCl, mg/L	Velocity, mL/min	Feed Solids, mg/L, $\underline{C}_o$	Filtrate Solids, mg/L, $\underline{C}$	$\lambda\underline{L} = \ln\left(\frac{\underline{C}_o}{\underline{C}}\right)$
			Titanium Dioxide		
4.6	0	83.3	97.6	54.8	0.577
4.0	0	93.4	95.2	45.6	0.736
3.5	0	88.0	102.4	3.2	3.466
3.0	0	87.7	102.4	0.0(0.8)	∞ (4.852)
2.5	0	93.7	98.4	0.0(0.8)	∞ (4.812)
2.0	0	95.2	94.4	3.2	3.384
4.9	500	52.6	89.2	3.2	3.328
5.2	1000	82.5	90.4	2.0	3.811
5.2	1500	90.9	88.0	3.2	3.314
5.3	2000	95.2	93.6	4.0	3.153
			Clay		
4.5	0	95.2	101.2	77.2	0.271
4.0	0	98.3	99.2	77.6	0.246
3.5	0	96.8	99.2	27.2	1.294
3.0	0	95.2	88.8	2.4	3.611
2.5	0	95.2	100.8	1.6	4.143
2.0	0	97.6	110.4	0.0(0.8)	∞ (4.927)
5.3	500	98.6	98.8	74.8	0.278
5.2	1000	94.8	96.0	90.8	0.056
5.3	1500	95.2	100.0	83.2	0.184
5.9	2000	105.0	95.6	74.4	0.251

Table II shows that as pH decreases, separation improves for both TiO_2 and clay. This is primarily due to the charge reduction or reversal on these particles. Thus, double layer repulsion is minimized, and as a result, particle-particle contact is enhanced. Also, attachment to sand grains may be improved at lower pH.

In the presence of sodium chloride, separation of TiO_2 is improved but that of clay is not. In general, the effect of electrolyte is to compress the double layer and, therefore, reduce the zeta potential. However, zeta potential is found to increase in the presence of an electrolyte. The reasons for this increase in zeta potential, also observed by others, are not clear. In the case of TiO_2 it appears that the improved separation in the presence of sodium chloride is due to reduction in double layer repulsion forces. The effect of a monovalent cation, like Na^+, in the separation of clay is small (26) due to the protective phosphate layer on clay particles. Clay particles are stabilized this way for ease in dispersion. The protective layer could easily be displaced by a trivalent cation like Al^{+++}, as will be seen in experiments with alum.

In the case of good separation, say when $\lambda \underline{L} \geq 3$ or $\underline{C}_o/\underline{C}_{\underline{L}} \geq 20$, double layer repulsion must be negligible. As a result, all collisions can be assumed to lead to attachment. In other words, $\alpha \cong 1$ when separation is good. From the relationships between λ and η developed in the last section, we have

$$\lambda = \frac{3}{2d_g} \eta \, \alpha(1-\varepsilon)$$

or in the case of good separation, $\alpha = 1$ and $\lambda \underline{L} \geq 3$,

$$\eta \geq \frac{2d_g}{L(1-\varepsilon)}$$

Therefore,

$$\eta \geq \frac{2 \times 0.02}{7.62(1-0.4)} = 0.00875$$

Thus, in the case of good separation, the collision efficiency, η, must be of the order of 0.01, i.e., one out of every hundred particles "seen" by a sand grain must collide with it.

It will be interesting to compute various contributions to the collision efficiency to determine the dominating transport mode. Results obtained from various theoretical or semitheoretical analyses are presented in Table III.

Table III shows that sedimentation or gravitational transport is negligible in the case of the TiO_2 and clay particles considered here. Also, contributions due to diffusion (assuming that particles follow Stokes Law) and due to interception (assuming that fluid velocity profile is parabolic) are negligible. Therefore, in the final addition, contribution by diffusion is obtained from the free surface cell model, and that by interception is obtained by assuming a flat velocity profile.

The collision efficiency for TiO_2 is somewhat lower than 0.00875 required for good separation. Therefore, we decided to modify the equations for collision efficiency. This was done by choosing different numerical constants in the expressions for η_D and η_I.

$$\eta_D = 10\left(\frac{kT}{d_g dU}\right)^{2/3} = 0.665 \times 10^{-2} \text{ for } TiO_2$$

$$= 0.289 \times 10^{-2} \text{ for clay}$$

and

$$\eta_I = \left(\frac{d}{d_g}\right) = 0.2 \times 10^{-2} \text{ for } TiO_2$$

$$= 0.7 \times 10^{-2} \text{ for clay}$$

Then, $\eta = 0.00874$ for TiO_2

and $= 0.01042$ for clay

TABLE III

THEORETICAL VALUES OF COLLISION EFFICIENCY

Mode of Transport and Assumptions	η, TiO_2	η, clay
1. Diffusion, η_D		
(a) Assume: Stoke's Velocity Profile		
$\eta_D = 0.9 \left(\frac{kT}{\mu d_g d U}\right)^{2/3}$	0.060×10^{-2}	0.026×10^{-2}
(b) Assume: Free Surface Equivalent Cell		
$\eta_D \doteq 3.43 \left(\frac{kT}{\mu d_g d U}\right)^{2/3}$	0.228×10^{-2}	0.099×10^{-2}
2. Sedimentation or Gravitational, η_G		
$\eta_G = \frac{\rho_p - \rho}{18\mu U} g d^2$	0.009×10^{-2}	0.053×10^{-2}
3. Interception, η_I		
(a) Assume: Parabolic Velocity Profile		
$\eta_I = 1.5 \left(\frac{d}{d_g}\right)^2$	0.001×10^{-2}	0.007×10^{-2}
(b) Assume: Flat Velocity Profile		
$\eta_I = 2 \frac{d}{d_g}$	0.400×10^{-2}	1.400×10^{-2}
Collision Efficiency $\eta = \eta_D^{(b)} + \eta_G + \eta_I^{(b)}$	0.637×10^{-2}	1.552×10^{-2}

Values of the various parameters are listed on the next page.

Values of Parameters Used in Table III

$\underline{k} = 1.38 \times 10^{-16} \frac{g\ cm^2}{sec^2\ °K}$

$\underline{T} = 298°K$

$\mu = 0.01 \frac{g}{cm\ sec}$

$\underline{U} = \frac{90\ (mL/min)}{\left(60 \frac{sec}{min}\right)\left(\frac{\pi}{4} \times (2.54)^2 cm^2\right)} \cong 0.3 \frac{cm}{sec}$

$\underline{d}_p$ - see Table I

$\underline{d}_g = 0.02$ cm

$\varepsilon = 0.4$

ρ_p - see Table I

$\rho = 1\ g/cm^3$

$\underline{g} = 981\ cm/sec^2$

It is clear from Table III or from the proposed empirical equations that both Brownian diffusion and interception are important modes of transport, whereas gravitational effects are negligible in the case of 1 μm size particles. In the case of latex particles with a density $\rho_p = 1.08\ g/cm^3$ and $\underline{d} = 0.5$ μm, one would expect that gravitational force can safely be neglected.

So far we have considered the case of good separation where every collision results in attachment, $\alpha=1$. Now, let us consider the case of TiO_2 and clay when their separations are not as good. In that case, from Table III,

$$\lambda L = 0.577 \text{ for } TiO_2$$
$$= 0.271 \text{ for clay}$$

Since collision efficiency depends primarily on hydrodynamic conditions, one can assume that it is the same as that for the good separation, i.e.,

$$\eta = 0.00874 \text{ for } TiO_2$$

$$= 0.01042 \text{ for clay}$$

It is possible to calculate the attachment efficiency for these particles from the relationship

$$\lambda = \frac{3}{2d_g} \eta \cdot \alpha \cdot (1-\varepsilon)$$

Since $\underline{L}$ = 7.62 cm, $\underline{d_g}$ = 0.02 cm and ε = 0.4,

$$\alpha = 0.192 \text{ for } TiO_2$$

$$= 0.076 \text{ for clay}$$

This means that only 20% and 8%, for TiO_2 and clay, respectively, of all collisions result in attachment. This can be attributed to double layer repulsion forces between these particles and sand grains.

Zeta potential can be used to measure the relative magnitude of charges on the particles. Both TiO_2 and clay have a negative charge of about -30 mv when dispersed in deionized distilled water. The difference in the attachment efficiency of the two particles may arise from the difference in the Van der Waals attractive forces. We know (27,28) that the magnitude of the attractive forces, due to London dispersion effect, is proportional to the Hamaker constant. In the case of water, it has been shown (9) that the Hamaker constant increases with the density of the particle. Thus, one would expect attractive forces to be larger for TiO_2 (density = 4 g/cm^3) than for clay (density = 2.5 g/cm^3). This explains, at least partly, the observed higher attachment efficiency for TiO_2 compared to that for clay. Quantitative analysis of repulsive and attractive forces is quite tedious and not very reliable. Therefore, this subject will not be pursued at this time.

Summarizing results obtained thus far, separation of TiO_2 or clay, dispersed in water, is not very good due to repulsive forces between these particles and sand grains. As the pH is lowered, particle charge and double layer repulsion decreases, and as a result separation improves. It is generally believed that increasing the ionic strength reduces double layer thickness and therefore improves separation. This is what happens in the case of TiO_2. However, in the case of clay, a protective phosphate layer significantly reduces the effect of monovalent cations on separation efficiency.

LATEX

Let us now consider some experiments with latex particles. Recall from Table I that these particles are 0.5 μm in diameter with a density of 1.08 g/cm^3. Thus, the size of latex particles is very close to that of TiO_2 particles (0.4 μm diameter). Results in Table IV show that the collision efficiency of latex particles is close to that of TiO_2 particles.

The fact that separation of latex particles is relatively poor even at pH 2 or at 5000 mg/L of NaCl, indicates that the attachment efficiency is small. In other words, the latex suspension is very stable. The zeta potential of a latex suspension is about -41 mv. This value changes to -11 mv at pH 2 and to -60 mv at 5000 mg/L of NaCl. Thus, latex particles are highly negatively charged, and this probably explains the observed poor separation. Also the lower density of latex may give rise to a relatively low magnitude of the attractive forces. Hence, one may conclude that compared to TiO_2 and clay, latex particles are difficult to separate by sand filtration.

Assuming that the empirical value of the collision efficiency, shown in the last column of Table IV, is reasonable, the collection efficiency α, for the

TABLE IV

FILTRATION OF LATEX-PARTICLES

pH	NaCl, mg/L	Velocity, mL/min	Feed Solids, $\underline{C}_o$, mg/L	Filtrate Solids, $\underline{C}$, mg/L	$\lambda\underline{L} = \ln\left(\frac{\underline{C}_o}{\underline{C}}\right)$	$\eta^a = \eta_D^{(b)} + \eta_G + \eta_I^{(b)}$	$\eta^b = \eta_D^{(e)} + \eta_G + \eta_I^{(e)}$
6.7	0	92.3	98.0	87.0	0.119	0.00696	0.00818
2	0	74.0	108.0	86.0	0.228	0.00727	0.00908
6.0	2000	105.0	96.0	88.0	0.087	0.00680	0.00771

[a] Collision Efficiency $= \eta = \eta_D^{(b)} + \eta_G + \eta_I^{(b)}$

$$= 3.45\left(\frac{kT}{\mu d_g d U}\right)^{2/3} + \frac{(\rho_p - \rho)g\, d^2}{18\mu U} + \left(\frac{d}{d_g}\right)$$

[b] $\eta = \eta_D^{(e)} + \eta_G + \eta_I^{(e)}$

$$= 10\left(\frac{kT}{\mu\, d_g dU}\right)^{2/3} + \frac{(\rho_p - \rho)g\, d^2}{18\mu U} + \left(\frac{d}{d_g}\right)$$

See Table I and parts of Table III for values of various variables.

latex particles and sand grains can be calculated.

$$\alpha = \frac{2\, d_g (\lambda L)}{3 \cdot L \cdot \eta (1-\varepsilon)} = \frac{2 \times 0.02 \times 0.119}{3 \times 7.62 \times 0.00818 \times 0.6}$$

or

$$\alpha = 0.0424$$

Thus the collection efficiency for the latex particles is much smaller than that for TiO_2 or clay particles.

As mentioned earlier, the collection efficiency is a balance between double layer repulsive forces and Van der Waals attractive forces. For the case of water, it has been shown (9) that these attractive forces are directly proportional to the particle density. For the TiO_2, clay, and latex particles considered here, the approximate relationship can be given by

$$\alpha = 0.04\rho$$

as shown in Fig. 5. Much more data with different types of particles are needed to get further insight into such a relationship.

TiO_2, CLAY, AND LATEX WITH ALUM

Filtration of colloidal particles is almost always carried out by pretreating the suspension so as to destabilize the particles. Although this can be done by adjusting pH or ionic strength, one of the commonest methods is to add a coagulant aid. We studied the filtration of TiO_2, clay, and latex particles in the presence of alum, and the results are shown in Table V. Two sets of experiments were carried out, one at pH 8.0 and the other at pH around 6.0.

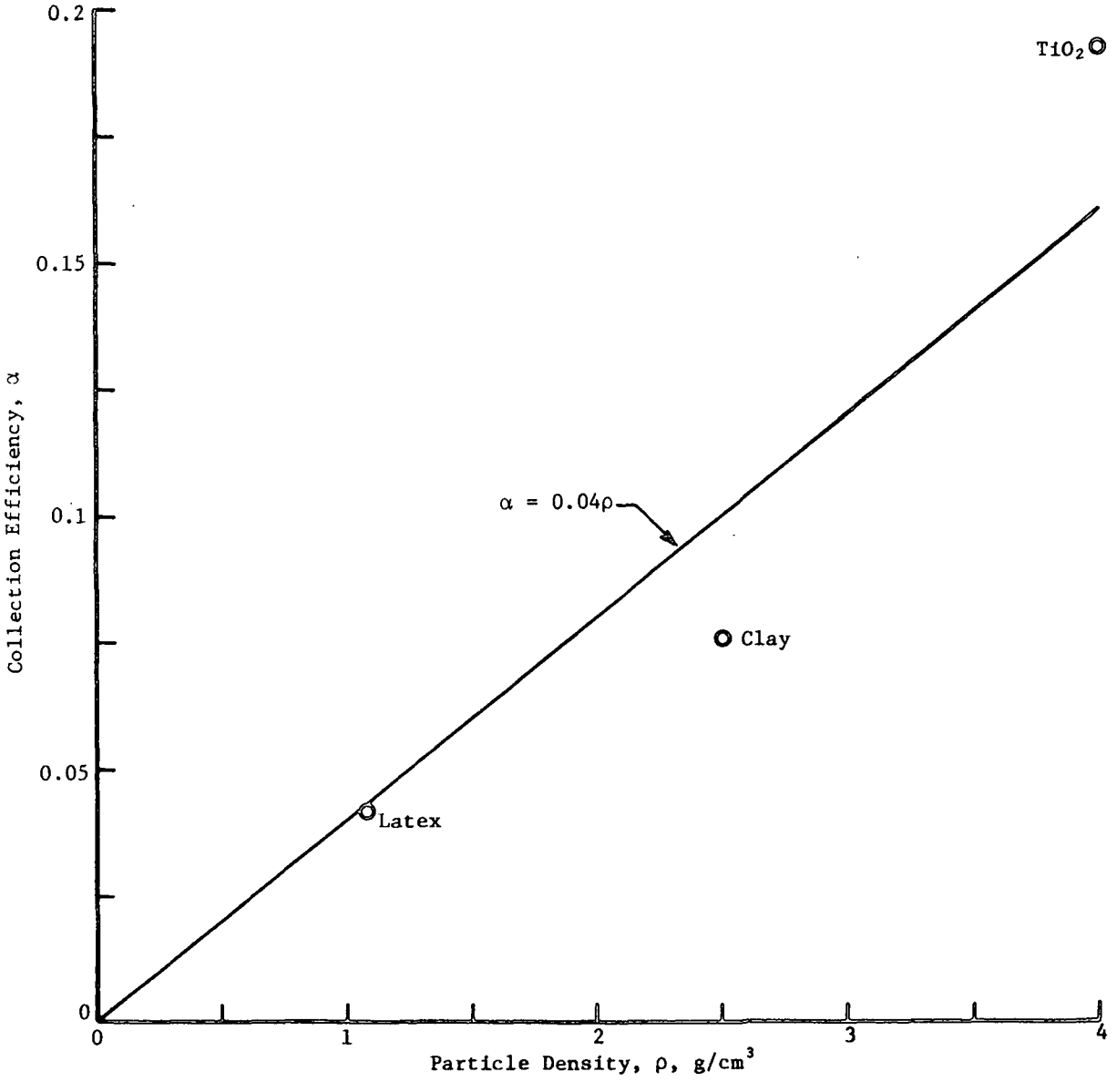

Figure 5. Collection Efficiency as a Function of Particle Density

TABLE V

FILTRATION OF TiO_2, LATEX AND CLAY

Feed	Velocity, mL/min	Feed Solids, C_o, mg/L	Filtrate Solids, C, mg/L	Efficiency, $\frac{C_o - C}{C_o} \times 100$	$\lambda L = \ln\left(\frac{C_o}{C}\right)$
I. Velocity of Water Through the Bed $\triangleq$ 90.0 mL/min					
TiO_2 + 25 mg/L alum pH 8	69.2	93	7	92.5	2.587
Clay + 25 mg/L alum pH 8	55.5	102	8	92.2	2.546
Latex + 25 mg/L alum pH 8	69.0	109	16	85.3	1.920
II. Velocity of Water Through the Bed $\triangleq$ 120 mL/min					
TiO_2 + 25 mg/L alum pH 5.9	97	92	19	79.3	1.577
Clay 25 mg/L alum pH 6.5	80	98	1	99.0	4.585
Latex 25 mg/L alum pH 5.8	102	97	4	95.9	3.188

The above two sets of experiments were made at different times, and consequently there is some difference in the rate of flow of water through the bed. In both cases the filtration rate is reduced by about 15 to 20% compared to the water flow rate. This is probably due to the removal of particles in the initial portion of the bed which will then offer extra resistance to the flow.

In all cases, separation efficiency is about 80 to 99%. Thus, it is clear that when particles are destabilized, separation efficiency is substantially increased. Since coagulation is a dynamic process, it is difficult to measure a representative particle size. It is, therefore, not possible to calculate collision efficiency in this case. Collection or attachment efficiency will be close to unity in most of the cases, since separation is generally high.

It is known that coagulation with alum (5) is effective between pH 5 and 8. In sand filtration also alum is effective in this pH range. Thus, simple jar tests can be used to determine effective dose and pH for coagulation prior to sand filtration.

SEPARATION OF MIXTURES

After considering the separation of a single component suspension, we studied the separation of mixtures of TiO_2 and clay, and of TiO_2, clay, and latex. Results are presented in Table VI. The weight of each component was kept approximately the same.

Table VI shows that the filtration of untreated mixtures (Experiment No. 1, 5) is poor, as observed in the case of a single component system. When particles are destabilized by lowering pH (Experiment No. 2) or by coagulating them with alum (Experiment No. 3), performance improves. The removal of latex particles in the presence of TiO_2 and clay at pH 2 (Experiment No. 6) appears to be better than in a single component system. However, one cannot be sure of this, since concentrations of individual components were not measured.

The interesting case is the separation of a mixture in the presence of sodium chloride. Recall that sodium chloride affected the filtration of TiO_2

TABLE VI

FILTRATION OF MIXTURES

Expt. No.	Feed	Velocity, mL/min	Feed Solids, C_o, mg/L	Filtrate Solids, C, mg/L	$\lambda L = \ln\left(\frac{C_o}{C}\right)$
1	TiO_2 ∿ 50 mg/L Clay ∿ 50 mg/L	98.7	100.0	82.0	0.198
2	TiO_2 ∿ 50 mg/L Clay ∿ 50 mg/L pH 2	71.4	95.0	0.0	∞
3	TiO_2 ∿ 50 mg/L Clay ∿ 50 mg/L 25 mg/L alum, pH 8	74.0	112.0	11.0	2.321
4	TiO_2 ∿ 50 mg/L Clay ∿ 50 mg/L NaCl ∿ 2000 mg/L	80.5	103.0[a] (45 mg/L TiO_2 58 mg/L clay)	38.0 (7 mg/L TiO_2 31 mg/L clay)	0.997
5	TiO_2 + clay + latex ∿ 50 mg/L each	71.0	150.0	109.0	0.319
6	TiO_2 + clay + latex ∿ 50 mg/L each pH 2	76.0	125.0	15.0	2.120
7	TiO_2 + clay + latex ∿ 50 mg/L each 2000 mg/L NaCl	70.0	146.0	88.0	0.506
8	TiO_2 + clay + latex ∿ 100 mg/L each 2000 mg/L NaCl	69.0	280.0 (TiO_2, 88 mg/L)	159.0 (TiO_2, 9 mg/L)	0.566
9	Filtrate from Expt. 8 pH 2	70.0	159.0 (TiO_2, 9 mg/L)	36.0 (TiO_2, 1.5 mg/L)	1.485

[a] Concentration of TiO_2 measured by TAPPI Method T 627 as 58 colorometric, Procedure: NB 3278 — p. 38.

(Table II) but not that of clay (Table II) and latex (Table IV). Consequently, when the mixture of TiO_2, clay, and sodium chloride was filtered (Experiment No. 4) most of the TiO_2 was retained, and most of the clay particles appeared in the filtrate. Similarly when the mixture of TiO_2, clay, latex, and sodium chloride was filtered (Experiment No. 7, 8), most of the latex and clay particles appeared in the filtrate, and TiO_2 was retained. The filtrate from this run, containing primarily clay and latex, was filtered again after the pH was adjusted to 2 (Experiment No. 9). From the results of Tables IV and II, one would expect that latex particles would appear in the filtrate while particles of clay would be retained. This is confirmed from the results in Table VI.

Thus, results of Table VI show that the filtration of mixtures follows, more or less, the behavior exhibited by the respective single component systems. Furthermore, it is possible to separate a mixture by preferentially filtering out appropriate components. Hence, sand filtration can be used as a solid-liquid, as well as solid-solid (in a liquid) separation. Not much work has been reported on the use of sand filtration for solid-solid separation. One may also note here that separation of TiO_2, clay, and latex particles is not achieved on the basis of their size differences but on the physicochemical nature of their surfaces and their interaction with sand grains.

THE EFFECT OF STARCH

Now we will consider filtration of colloidal particles like TiO_2 and clay in the presence of starch.

Experiments were carried out with three forms of starch:

(1) Pearl starch — unmodified cornstarch containing about 25% amylose (straight chain polymer) and 75% amylopectin (branched chain polymer)

(2) Amylon II — 65% amylose and 35% amylopectin

(3) 100% Amylopectin

A 4% starch solution was cooked at 94°C for 25 minutes with moderate agitation. The solution was then diluted to about 1.6% with warm deionized distilled water and allowed to cool slowly at room temperature. A fresh solution was cooked each day.

Results of the starch-TiO_2 and starch-clay filtration are presented in Table VII. These results should be compared with those obtained without starch which are also given in Table VII.

Filtration velocity is somewhat reduced when Amylon-II or 100% amylopectin is present, and it is markedly reduced in the presence of pearl starch. In some cases, particularly with TiO_2 and starch, a slime layer was visible at the surface of the filter bed. It is interesting to note that the presence of starch in the stock slurry is known to reduce the drainage rate or freeness.

Separation efficiencies without starch, with pearl starch, or with Amylon-II are not too different. However, 100% amylopectin seems to reduce the separation efficiency of TiO_2 at pH 2 and at high ionic strength (2000 mg/L NaCl). It may be possible that adsorption of hydrophilic starch on TiO_2 particles may occur under these conditions and consequently retard their separation. Coating clay is not so affected, probably due to the protective phosphate layer. More work in this area is needed to better understand the effect of starch on sand filtration.

TABLE VII

FILTRATION OF COLLOIDAL PARTICLES AND STARCH

Feed	No Starch			Pearl Starch, 21 mg/L			60-70% Amylose, 21 mg/L			100% Amylopectin, 21 mg/L		
	Velocity, mL/min	Feed Solids, C_o mg/L	Efficiency, $\frac{C_o - C}{C_o} \times 100$	V, mL/min	C_o, mg/L	Efficiency	V, mL/min	C_o, mg/L	Efficiency	V, mL/min	C_o, mg/L	Efficiency
TiO_2												
As is	83.3	97.6	44	6.8	89	36	34.6	99	44	19.3	92	78
pH 2	95.2	94.4	97	27.0	80	95	40.0	106	100	58.8	90	59
2000 mg/L NaCl	95.2	93.6	96	20.0	83	100	30.3	97	94	45.1	76	53
25 mg/L Alum, pH 8	69.2	93.0	93	12.1	115	100	60.6	99	94	16.2	114	100
Clay												
As is	95.2	101.2	24	16.3	104	38	54.0	103	35	56.0	108	52
pH 2	97.6	110.4	99	50.0	137	89	74.0	112	87	48.8	88	93
2000 mg/L NaCl	105.0	95.6	22	28.1	117	49	62.5	102	63	52.6	92	26
25 mg/L Alum, pH 8	55.5	102.0	92	5.0	134	93	18.5	126	91	5.0	--	--

CONCLUSIONS

The following conclusions can be drawn from the sand filtration results presented in this report:

(1) Separation Mechanisms: The principal modes of transport for the removal of about 1 μm size colloidal particles (like TiO_2, clay, and latex) appear to be Brownian diffusion and interception. Gravitational transport (also known as sedimentation), negligible in this case, may become important for particles greater than about 5 μm.

(2) Separation Efficiency: Separation of TiO_2, clay, and latex particles by media filtration is poor primarily due to double layer repulsive forces. However, separation efficiency increases significantly when these particles are "destabilized" (i.e., charge on the particles is reduced). Depending on the nature of the particles, destabilization can be effected by adjusting pH or increasing ionic strength or adding a coagulation aid, like alum. Addition of alum or other coagulating aid is the most common and effective method of improving separation of colloidal particles.

(3) Filtration of Mixtures: Particle-particle interaction is minimal in dilute systems considered here. Therefore, the performance of mixtures can, in general, be predicted from the corresponding single component systems. It is also possible to preferentially destabilize and, hence, separate one or more component(s) from a mixture.

(4) The Effects of Starch: The presence of starch reduces the filtration velocity significantly by forming a gel layer or cake in the top layers of the bed. The behavior of separation efficiency is not entirely clear and needs more work.

FUTURE WORK

Granular media filtration seems to have potential as a postsaveall unit. Biosolids from secondary effluents could be removed by granular media filtration. Therefore, the effectiveness of media filtration in polishing clarified water from saveall units and biological treatment systems will be investigated in the coming year.

Another important method for the removal of suspended solids is by dissolved air flotation. In this process, separation is achieved by attachment and/or entrapment of air bubbles. Relative importance of these mechanisms and the role of chemicals in enhancing one or the other separation mechanisms will be investigated.

Our work on granular media filtration and dissolved air flotation will be discussed in the next progress report. Our future plans include a study of microscreening and sedimentation processes.

ACKNOWLEDGMENTS

Special thanks to Mr. Tony Wisneski and Mrs. Jacquelyn Kern for conducting experiments and analyses. Discussions with Mr. John W. Swanson and Dr. Lynden J. Stryker were quite helpful.

NOMENCLATURE

A	cross-sectional area of the bed, cm^2
A_D, A_I, A'_I	numerical constants
C	particle concentration, mg/L
C_o	particle concentration in a stream entering the bed, mg/L
C_L	particle concentration in a stream leaving the bed, mg/L
ΔC	change in concentration as the fluid passes over a bed of thickness ΔZ, mg/L
D	diffusion coefficient, cm^2/sec
d	particle diameter, cm
d_g	diameter of the sand grain, cm
g	gravitational acceleration, cm/sec^2
K	Kozeny-Carman constant, cm^2
k	Boltzmann constant = 1.38×10^{-16} g cm^2/sec^2 °K
L	length of the bed, cm
Pe	Peclet Number = $U\ d_g/D$
ΔP	pressure drop across the bed
T	absolute temperature, °K
U	filtration velocity, cm/sec
V	volumetric filtration rate, cm^3/min
Z	coordinate measured from the entrance of the bed, cm
ΔZ	infinitesimal thickness of the bed, cm
α	collection efficiency
ε	porosity of the bed
η	collision efficiency
η_D	collision efficiency due to Brownian diffusion

η_G	collision efficiency due to sedimentation
η_I	collision efficiency due to interception
λ	filtration coefficient, cm^{-1}
μ	fluid viscosity, g/cm sec
ρ	fluid density, g/cm^3
ρ_p	particle density, g/cm^3

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Mahendra R. Doshi

Mahendra R. Doshi, Project Leader
Research Fellow
Systems Engineering Group
Environmental Sciences Division

Peter E. Parker

Peter E. Parker
Research Associate-Group Leader
Systems Engineering Group
Environmental Sciences Division

Approved by:

Hardev S. Dugal

Hardev S. Dugal
Director
Environmental Sciences Division